



Mercury Cycling in the Hudson/Raritan River Basin

by

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I. INTRODUCTION

The purpose of this study is to estimate all major flows of mercury – from environmental and anthropogenic sources – within the drainage basins of New York/New Jersey Harbor (Figure 1). Our goals are to provide: 1. the proper magnitude for the mercury reservoirs and flows relating to NY/NJ Harbor, and 2. a firm scientific basis for assessing the scale and impact of pollution-related mercury. The current database is inadequate for more refined approaches or simulations. Thus, reasonable assumptions and simplifications were necessary, accounting for weaknesses in the available biogeochemical and environmental information for the study region. Our approach provides a first-order estimate for a mass balance of mercury sources, transport and deposition within NY/NJ Harbor. Although there are uncertainties associated with our findings, the estimated mass balance constitutes a useful framework for more precise assessment of mercury sources, cycling, behavior and fate in the Harbor.

II. METHODS

Direct measurements or models of most mercury fluxes are insufficient for aggregate estimates over entire drainage basins. Conventional measurement approaches often, in the aggregate, yield biased and incomplete quantities which can not be validated. However, these fluxes can be estimated from economic and population data, process data, and the mass-balance principle. Mass balances are used to validate, or detect inaccuracies, in our analyses. We emphasize “top down” methods to estimate regional mercury pools and fluxes. “Bottom up” sums of many individual activities or facilities are often less reliable. This “industrial ecology” approach is now widely recognized as valuable. One of the earliest applications of industrial ecology provided estimates of anthropogenic pollutant emissions (including mercury) in the Hudson River basin from 1880 to 1980.¹ Where data are available, we have calculated flux estimates for mercury over the last three decades.

The present draft presents preliminary regional mass balances for Hg in the atmosphere, land, watersheds, and NY/NJ Harbor. These budgets are used to test and constrain estimates of the behavior and fate of mercury in the study region (i.e., Hudson/Raritan Basin). This information is presented in the form of summary tables and figures complemented with supporting data, discussion, and initial inferences or conclusions.

¹ R.U. Ayres and L. W. Ayres (1999), *Accounting for Resources*, vol. 2 (Cheltenham, UK: Edward Elgar), ch. 5.

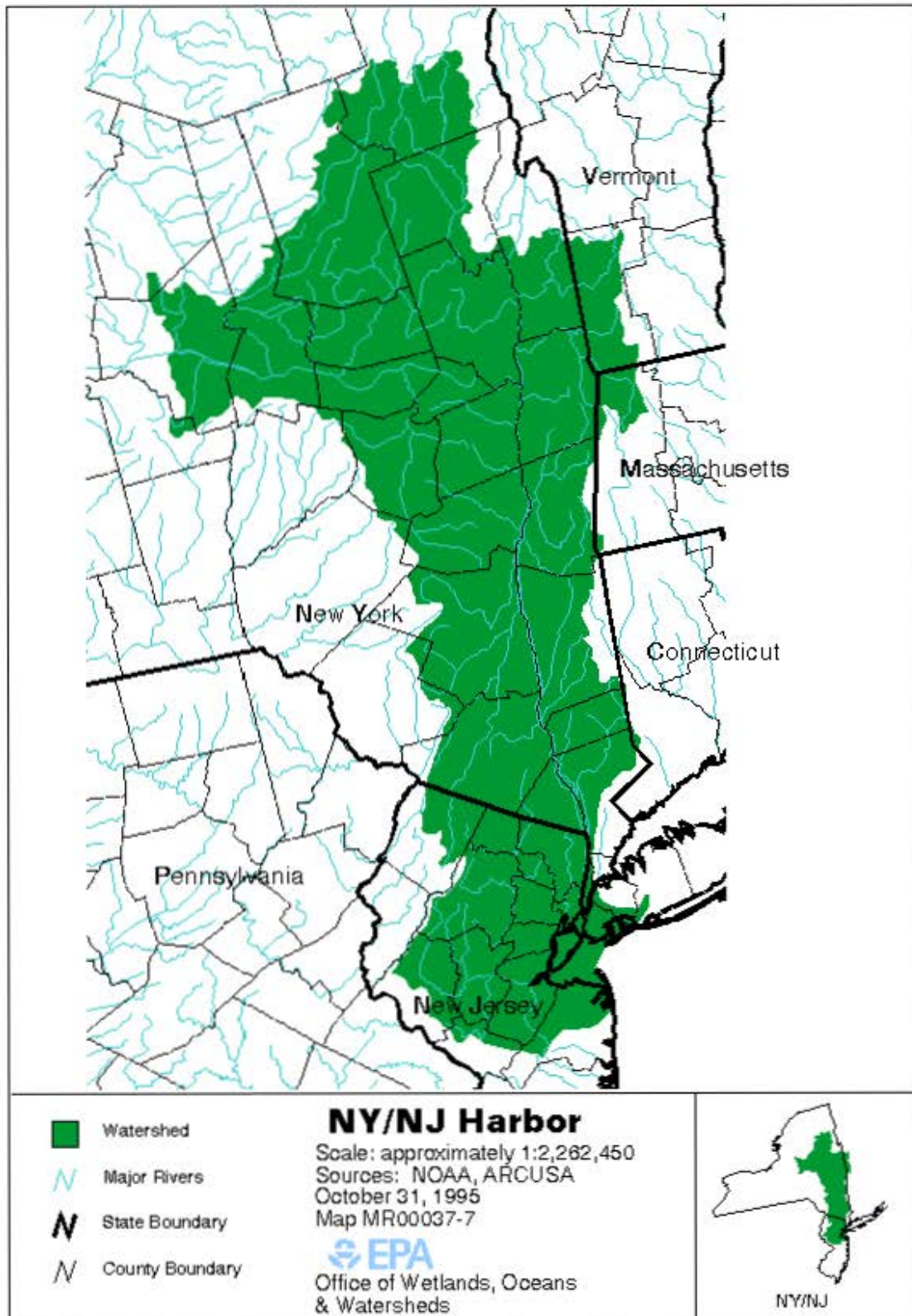


Figure 1. New York/New Jersey Harbor Drainage Basins.

III. MERCURY SOURCES / CONTRIBUTIONS

Some uses of mercury are directly proportional to population in the study area: healthcare activities, and consumption and disposal of consumer goods. All these uses depend upon the demand for goods and services by the resident population, hence are proportional to population size. Population size is widely used as a surrogate for mineral consumption.²

The U.S. resident population in 1980, 1990 and 2000 is available from the U.S. Census Bureau. This Bureau has provided us with reliable, national and county census data. The regional population within the study area was estimated by summing the populations of counties wholly or partly within the drainage basins. For counties not entirely within the drainage basins, county contributions to the regional total were assumed to be proportional to the fraction of the county within the basins.

National domestic consumption of mercury for all purposes has declined since the early 1970s by an order of magnitude. Many industrial uses have been eliminated each year in favor of less toxic substances.³ Mercury cell chlor-alkali manufacturing has been a major source of industrial mercury loss nationally. However, there has not been any industrial preparation of chlorine and caustic soda within the study area since the 1980s.⁴

Table 1 presents total industrial mercury use and total industrial use minus national use for chlorine and caustic soda manufacturing. The net national usage, adjusted for regional population, is the most appropriate, robust basis for estimating total mercury usage by industry in the study area. That is: regional industrial mercury usage = adjusted national industrial usage • (regional population / national population). Alternative estimates based upon summing use by individual industries would probably contain more significant bias. See for example Ayres and Ayres (1999), Guinee *et al.* (1999), and Sunderland and Chmura (2000). Additional industrial uses of mercury include manufacturing of fluorescent and high-intensity-discharge lamps, thermometers of several kinds, measuring and control instruments, thermostats, switches and relays, catalyst in production of organic chemicals, and dental amalgams.⁵

Table 1.
Mass of Mercury Used by Industry within
the Hudson/Raritan Drainage Basins Over 20 Years

Mercury Use and Consumption	1980	1990	2000
U.S. resident population (1,000s) ⁶	248,710	250,567	274,024
Hudson/Raritan basin population (1,000s) ⁷	14,832	14,752	15,170
U.S. industrial use of mercury (1,000 kg) (A) ⁸	2,033	720	346
National use of mercury for manufacturing chlorine and caustic soda (1,000 kg) (B)	326	247	154
A – B (1,000 kg)	1,706	473	192
Regional industrial use of mercury (1,000 kg)	102	27.8	10.6

² E.M. Sunderland and G. Chmura (2000), "An Inventory of Historical Mercury Emissions in Maritime Canada: Implications for Present and Future Contamination," *Sci. Tot. Env.* 256: 39-57.

³ U.S. Bureau of Mines, *U.S. Geological Survey*.

⁴ Ayres and Ayres, *Accounting for Resources*, 2.

⁵ The draft paper by Julio Huato on *Estimation of the Level of Activity of Mercury-Consuming Industries in the New York/New Jersey Harbor Watershed* provides detailed information on industrial uses of mercury.

⁶ U.S. Census Bureau, Population Division, Population Estimation Program, July 2000. Downloaded from <http://www.census.gov/population/estimates/county>. 1980 values from published U.S. Census Bureau General Population Characteristics.

⁷ U.S. Census Bureau, "County Estimates for July 1999" used for 2000 estimates. Downloaded from <http://www.census.gov/population/estimates/county>.

⁸ 1980 and 1990 U.S. Bureau of Mines, Minerals Yearbook; Feb 1998; Feb 2000 U.S. Geological Survey, 1993-1996, 1994-1998. Minerals Yearbook, Minerals and Materials Information. From the CD Rom.

A. Carbon Combustion

This section on carbon combustion benefits greatly from estimates derived from the important work by the New Jersey Mercury Task Force. For all forms of carbon we assume that mercury releases are direct functions of carbon burned (expressed as Btu), and that carbon burned is a direct function of population. Below are estimates for releases from residual oil, coal, natural gas, distillate fuels and gasoline, which are summarized in Table 2.

Residual oil (number 6 fuel oil) is composed of the heaviest components of crude oil. It is that portion of crude oil which is left, after all other products are removed, hence the name residual. Mercury, as a contaminant, is emitted to the atmosphere from burning residual oil. The New Jersey Task Force estimated that 22 pounds of mercury are liberated per 2.3 million barrels of residual fuel oil burned. It is assumed that all mercury present in residual oils is released to the atmosphere during the combustion process. Residual oil is consumed in large boilers at utilities, commercial and industrial facilities.

Table 2.
Estimated Mass of Mercury (kg/y) Emitted from Combustion of Different Fuels

Source of mercury	1980	1990	1997/2000*
Coal	613 ± 244	686 ± 273	611 ± 243
Distillate fuels	10	10	8
Gasoline	36	38	38
Natural gas	--	--	--
Residual fuel oil	109	110	113
Wood	?	?	?
* Estimates are for 1997 or 2000, depending upon data availability.			

In 1997 the study area population within New Jersey (4,642,000) was 57% of the New Jersey population, and the study area population in New York (10,372,000) was 57% of the New York population. In 1997 the State of New Jersey burned 58.8 trillion Btu⁹, releasing an estimated 88 pounds of mercury from combustion of residual oil. Scaling up New Jersey's 1997 combustion of 58.8 trillion Btu to 167.2 trillion Btu for the entire study area, yields 250 lb (113 kg) of mercury on combustion.

In 1990 the study area population within New Jersey (4,475,000) was 58% of the New Jersey population, and the study area population in New York (10,247,000) was 57% of the New York population. Population change between 1990 and 1997 was (-) .00979%, yielding 110 kg of mercury during 1990 on combustion of residual oil within the entire study area.

In 1980 the study area population within New Jersey (4,385,000) was 60% of the New Jersey population, and the study area population in New York (10,447,000) was 59% of the New York population. Population change between 1980 and 1990 was (-) .00742%, yielding 109 kg of mercury on combustion of residual oil within 1980.

Coal is burned predominantly by utilities for electric power generation. Coal cleaning reduces the amount of ash produced, thereby reducing the amount of mercury emitted. However, the quantity of mercury removed varies primarily with the type of coal burned: anthracite, bituminous, sub-bituminous, or lignite.¹⁰

During the 1990s about 320 ± 128 kg of mercury was emitted to the atmosphere during coal combustion throughout New Jersey.¹¹ Scaling New Jersey's population to that of the Hudson-Raritan Basin, yields 611 ± 243 kg of mercury per year in the 1990s. Relatively small temporal changes in coal burned within the

⁹ USDOE, *State Energy Data Reports* (1997).

¹⁰ US EPA (2000), *The Use and Release of Mercury in the United States* (Washington, D.C.: Office of Research and Development).

¹¹ New Jersey Mercury Task Force (2001).

study area¹² indicate that about 613 kg of mercury were emitted in 1980, 686 kg in 1990, and 611 kg in 1997 (Table 2).

Natural gas is used in utility and industrial boilers and combustion turbines, as well as in home and commercial water heaters. Natural gas contains only trace quantities of mercury.¹³ The 1997 EPA Mercury Report to Congress estimates that national emissions of mercury from utility boilers burning natural gas is only four pounds per year. This quantity scaled to the Hudson-Raritan study area is negligible (Table 2).

Distillate fuels include jet fuels, diesel fuels, heating oil, and kerosene. Mercury is a contaminant in all distillate fuels. All the mercury present is released to the atmosphere during combustion.

Within the study area about 508 trillion Btu of distillate fuels were burned in 1980, about 518 trillion Btu in 1990, and 421 trillion Btu in 1997.¹⁴ The U.S. EPA (1997) estimated that 7.2 pounds of mercury are emitted per 10¹² Btu of distillate oil combusted. However, based upon recent data (obtained using ultra-clean sampling and analysis techniques) this emissions estimate is unrealistically high. The mass of mercury was only about 10 pounds in all distillate fuels burned in New Jersey during 1997¹⁶. Scaling this quantity of distillate fuels from the NJ population to the population of the study area yields 18 to 23 pounds (8 to 10 kg) of mercury released to the atmosphere within the Hudson-Raritan basin (Table 2). (Obviously, some large fraction of the jet fuel sold regionally is consumed globally, whereas unidentified quantities of jet fuel are imported from elsewhere and burned locally.)

Gasoline is consumed in large quantities within the study area by a variety of automobiles, trucks, boats, and small engines. Recent analyses of mercury in gasoline have shown consistently low concentrations, in the range of 1-3 ppb.¹⁵ This is equivalent to 0.45 kg of mercury in 7.8 trillion Btu, or 10¹⁰ kg of gasoline. Scaling by population, the mercury emitted from gasoline burned within the study area is 36 to 38 kg of mercury per year (Table 2). Because of the limited data on mercury emissions from gasoline, these estimates are highly uncertain.¹⁶

The quantities of mercury in **wood** burned for residential use and on industrial scales (e.g., burning sawdust and wood chips) are quite variable. Existing data on wood burning do not seem adequate to reliably estimate mercury releases in the study area. The emissions are probably not minor but are less than the emissions of each carbon-based fuel.¹⁷

B. Municipal Solid Waste

Municipal Solid Waste (MSW) is generated by residential, commercial and institutional sources, in both rural and urban areas. MSW does not include solid waste that cannot be compacted such as tree trunks, appliances, mattresses, construction/demolition waste and tires. A small fraction of the mercury in MSW is recycled. The remainder of MSW is disposed of in landfills, on the ground, or incinerated. Most of the mercury from incinerated MSW is initially vaporized regardless of chemical species. However, as the combustion gases cool, mercury species and condensates can attach to the "flue dust" in the incinerator. This precipitate is deposited in landfills.

Some elemental and reactive gaseous mercury escapes the collection system and is emitted to the atmosphere. Reactive gaseous species of mercury have short atmospheric lifetimes, and will deposit

¹² USDOE, *State Energy Data Reports* (1997).

¹³ L. Phannenstiel, C. McKinley, and J. Sorenson, "Mercury in Natural Gas," presented at American Gas Association, Operation Section Transmission Conference (Las Vegas, 76-T-12), p. T-202.

¹⁴ USDOE, *State Energy Data Reports* (1997).

¹⁵ L. Liang, M. Horvat, and P. Danilchik (1996), "A Novel Analytical Method for Determination of Picogram Levels of Total Mercury in Gasoline and Other Petroleum Based Products," *Science for the Total Environment* 187: 57-64.

¹⁶ New Jersey Mercury Task Force (2001).

¹⁷ US EPA 2000; Sunderland and Chmura, "An Inventory of Historical Mercury Emissions."

locally. Most of the elemental mercury enters the global cycle and is dispersed broadly.¹⁸ For scale, the state-of-the-art SEMASS incinerator at Rochester, Mass, processes one million tons of MSW annually and emits to the atmosphere 34 kg Hg. The flue dust represents 7.7% of the feed and on the average contains 18 parts per million of mercury (1,400 kg).¹⁹ Based upon the mass of mercury collected in the SEMASS flue dust and the amount emitted to the atmosphere, Themelis (2000) estimates that the SEMASS feed of MSW contains about 1.6 ppm of mercury -- which is close to the estimate for NJ MSW, as noted below.

New Jersey's MSW contains an estimated 2 ± 0.5 ppm of mercury.²⁰ New Jersey's five MSW incinerators are required to report mercury concentration in the stack gas at least once a year. These measurements show a consistent and dramatic decline in annual mercury emissions (from 2,000 kg/y to 150 kg/y) over the past decade. These reductions resulted primarily from installation of carbon-injection pollution-control systems. Mercury concentrations in the combusted MSW have probably decreased by only one ppm or so since the early '90s²¹ (New Jersey Mercury Task Force 2001).

The New Jersey Mercury Task Force also estimated that the 1999 mercury emissions to air from the two New Jersey incinerators in the study area (in Essex and Union Counties) were about 200 lbs (= 90kg Hg).²⁴ This number accords reasonably well with the 34 kg of Hg/year emissions from the million tons of solid waste processed by the SEMASS incinerator. The two New Jersey incinerators burn about 1,350,00 tons of waste per year.²⁴ At 2 ppm of mercury, this waste contains about 2,500 kg mercury. Hence, these two incinerators emit to the air about 3.5% (90 kg /2,500 kg) of the mercury in the waste.

The mass of MSW generated within the study area in 1999 has been tabulated by state.²² We have adjusted these state-generated values by adding the quantities imported and subtracting the quantities exported. The reported mass of MSW discarded within the study area in 1999 was 18.76 M (short) tons (3.9 M tons in New Jersey, 14.6 M tons in New York, and 0.26 M tons in Berkshire County, MA). We estimated the total mass of waste incinerated in the study area at 2.15 M tons, in six incinerators (two New Jersey facilities, three in New York, and one in Massachusetts).²³

The MSW combusted in the Hudson-Raritan Basin is expected to contain, on average, 2 ppm of mercury, as in New Jersey. So, the amount of mercury in the feed to study-area incinerators was 3,900 kg (2.15 M tons MSW x 2 ppm Hg x 1,000/1.1), while the percentage emitted to the atmosphere is expected to be the same as measured in New Jersey (i.e., 3.5%). Thus, the mercury emitted to air from incinerators in the study region in 1999 was about $3,900 \text{ kg} \times .035 = 140 \text{ kg}$.

As noted above, the total MSW disposed within the Hudson-Raritan Basin in 1999 was 18.76 M tons, of which 2.15 M tons were incinerated. Therefore, the amount placed in landfills was about 16.6 M tons. Assuming the concentration of Hg in the study area MSW was 2 ppm, the landfilled material in 1999 contained about 34,000 kg of mercury (16.6 M tons MSW x 2 ppm Hg = 33.2 tons or 30,200 kg plus 3,760 kg Hg (3,900 – 140 kg) attached to flue ash from incineration (i.e., about 34,000 kg Hg total).

Current measurements on certain state- and EPA-regulated landfills indicate effective sequestration of mercury for the short term.²⁴ However, there is no evidence about what happens to the mercury long after landfills are closed. Long-term emissions of liquid and gas from old landfills are expected. Mercury will eventually be released to the atmosphere, surface- and groundwater. Therefore, long-term cumulative mercury release from landfills could be two orders of magnitude greater than the short-term atmospheric release from incineration.

¹⁸ Lamborg *et al.*, (2000).

¹⁹ Themelis and Kim (2000).

²⁰ M. Aucott, Personal communication (2000).

²¹ M. Aucott, personal communication.

²² Goldstein, N. (2000).

²³ U.S. EPA, (2000). Office of Air and Radiation, An Inventory of Anthropogenic Mercury Emissions in the United States. Vol 2.

²⁴ New Jersey Mercury Task Force, (2001); M. Aucott, personal communication.

Recent yearly mercury emissions from coal-fired power plants, incineration, and the potential long-term release from landfilled MSW are summarized in Table 6. It is important to note that total environmental loadings from these sources are much larger than indicated by the 1999 emissions. Mercury-containing wastes have been discarded in the Hudson-Raritan Basin for over one hundred years, and the annual discharges of mercury were very large until the 1990s.

C. Industry

Environmental losses of mercury during industrial processes have declined substantially over time. This is due to a sharp drop in consumption by industry (Table 1), increased regulation of mercury wastes, and improved technologies for minimizing mercury wastes.²⁵ Current industrial process losses are so small they are not even good indicators of historical emissions. However, the decreasing use and waste of mercury has not led to comparable declines in environmental contamination. Millions of tons of mercury have already been lost to Hudson/Raritan environments since 1880,²⁶ much more than regional industry now emits over decades (see Tables 1 and 6). Residuals of these historical emissions still constitute major non-point mercury sources. For nearly 200 years these residual wastes have been dispersed widely from their original industrial sites. Dispersal via water to New York/New Jersey Harbor and coastal areas has been facilitated by local sea-level rise and resulting tidal exchange over large waste areas.

We have not tried to quantify emissions by industry or by product. This was not necessary for our purposes, and such detailed accounting would have been exceptionally labor-intensive. Others have compiled direct measurements of mercury concentrations in some consumer products, e.g.²⁷ EPA's Toxic Release Inventory (TRI) data were not used because of well-known biases²⁵.

Mercury is extremely mobile because of its high vapor pressure and low affinity for oxygen. Recognizing these things, Ayres and Ayres point out that:

Deposition in landfills or sediments is no guarantee of long-term immobilization....
Remobilization from virtually any use or waste is only a question of time. Thus, it seems likely that virtually all the mercury that has been mined in the past has been, or will be, released eventually.²⁸

This is a critically important observation for New York/New Jersey Harbor. Several of the world's largest industrial users, emitters, and processors of mercury were located on the margins of the Harbor for nearly 100 years before environmental dangers of mercury were recognized in 1970.²⁹ Plants were built to process or reclaim large mercury waste deposits from these industries. "Mercury cell" chlor-alkali plants were also built by GAF, Inc. in Linden, NJ and by Maquite, Inc. in Elizabeth, NJ. The mercury cell process was in commercial use by 1894. This process required large quantities of mercury in the manufacture of chlorine and sodium hydroxide (caustic soda) by electrolysis of sodium chloride (brine).³⁰ Mercury losses to the environment were about 300mg/kg of chlorine produced. This became the dominant, very leaky, process for producing chlorine and caustic soda by the early 1900s. Most chlor-alkali plants built from 1920 to 1950 used diaphragm cells rather than mercury cells. A new mercury cell design was commonly used after 1950, using mercury cathodes and inert anodes. Bathed in brine, a current flows to the mercury cathode where sodium ions amalgamate with the mercury. Metallic mercury on the cathode is then recycled, but even today mercury emissions probably remain about 150mg/kg of product. The mass of mercury lost to water and to air is roughly similar. Additionally, some mercury is lost in brine purification, some accumulates on pipes, pumps and valves of the plant, and – most

²⁵ Ayres and Ayres, ch. 5.

²⁶ Ayres and Ayres, ch. 8.

²⁷ Association of Metropolitan Sewerage Agencies. (2000).

²⁸ Ayres and Ayres. *Accounting for Resources*, 2. p. 160.

²⁹ Ibid, Ch. 8.

³⁰ Ibid. Ch. 5.

significantly – water treatment muds and sludges. Apparently, more than twice the reported national usage of mercury by the chlor-alkali industry was accumulating in waste sludges until 1992. Disposition of these sludges was not known, until 1992 when EPA expanded its Toxics Release Inventory (TRI) reporting to include “source reduction and recycling.” Regionally, most of these highly-contaminated sludges were dumped in landfills or on the margins of New York/New Jersey Harbor. Beginning in 1992, these sludge accumulations became illegal, and chlorine producers had to begin recycling.³¹

One of the largest industrial uses of mercury from the turn of the century through most of the 1980s was battery manufacture. One major battery manufacturer was located at Silverlake, NJ. Mercuric nitrate was widely used in the manufacture of felt hats. Many hat manufacturers were located in or near Newark, NJ and New York City. Mercuric fungicides were produced by several small firms in northern New Jersey.³⁰ Many additional mercury-using and -recycling firms were also sited around the Harbor, including the Troy Chemical Corp., Newark, NJ (originally [1892-1931] owned by Heller and Merz Company).

Environmental concentrations of mercury and other chemicals are still being quantified at many contaminated sites within the Raritan watershed by the New Jersey DEP Site Remediation Program. (Sediments and groundwater at many of these sites also remain contaminated with lead, arsenic, chromium, benzene, tetrachloroethene, naphthalene, and other organic and inorganic chemicals.) The DEP Site Remediation Program is also attempting to estimate modern fluxes of mercury from contaminated sites to the Harbor. Contaminated sites in New York State have been conveniently portrayed by the New York Public Interest Research Group (NYPIRG) at <http://www.nypirg.org>. Although NYPIRG shows all NYS Superfund Sites, enough information is provided to assess sites contaminated with mercury. Direct estimates of mercury flux from all these sites to the Harbor, its watershed, groundwater and atmosphere would be very expensive at the least, and may not be quantitatively feasible. In any case the largest, most relevant, fluxes almost certainly occurred from 10 to over 100 years ago.

GAF, Inc. (now International Specialty Products) in Linden, New Jersey functioned as a large chlor-alkali plant from 1919 to 1991. The 147 acre site is entirely filled with dredged material, and abuts the Arthur Kill near Prall's Island. Mercury concentrations in soils nearly reach 2,000 mg/kg. Ground water concentrations vary, but in 1996 were 420-1600 µg Hg/l in two shallow wells. These ground water concentrations increased by factors of >2 to 168 from 1991 to 1996. Large volumes of contaminated ground water (>32,000 gal/d) flow under the site to the Arthur Kill and Piles Creek (which also flows into the Arthur Kill).³²

LCP Chemicals, Inc. is located immediately to the south of the GAF site on 26 acres of filled wetland. From 1972 to 1985 the LCP facility was used to produce chlorine using the mercury cell electrolysis process. The site is drained by South Branch Creek which discharges into the Arthur Kill. GAF, Inc. operated a chlor-alkali plant on this site since 1952. Metallic mercury in brine from the mercury cell process was partially recovered and recycled. The remaining mercury containing sludge was placed in a 23,000 m³ brine sludge lagoon on the property, operated by LCP. As part of the LCP operation, lagoon supernatant was piped to a wastewater treatment system prior to being discharged to South Branch Creek. Major releases of brine sludge were partially documented from 1974 until legally imposed closure of the lagoon in 1984. Sludge accumulated in this lagoon for more than 20 years before its closure. Lagoon closure entailed sludge dewatering, compacting, capping with a two-foot layer of clay, and monitoring for mercury and other toxicants released to the environment. Concerns persist over mercury wastes generated, stored, or disposed of from this facility over 33 years (mercury contaminated sludge, mercury vapors, contaminated process wastewater, contaminated soil and ground water, wash-down fluids, and stormwater runoff). The firm had several environmental discharge violations from 1975 to 1985: e.g., mercury in brine and sludge. In 1982 surface soil samples from the perimeter of the brine sludge lagoon ranged from 27 to 1,580 ppm. Water from test wells ranged from 36 to 772 ppm Hg, with concentrations generally decreasing with depth. Mercury concentrations at 40-47 feet ranged up to 5 ppm. Concentrations of mercury measured by an EPA Site Inspection in 1995 – 10 years after production had ceased -- were 93 ppb in waters of the LCP outfall,

³¹ Ibid. Ch. 8.

³² March 24, 2000 letter from Ian R. Curtis, New Jersey DEP to Carl R. Eckart, International Specialty Products, Wayne, NJ.

~1,060 ppm in sediment at the LCP outfall, and ~350 to ~470 ppm in sediments downstream of the brine sludge lagoon. High concentrations of additional chemicals were also measured (e.g., arsenic, cadmium, zinc, silver).^{33,34}

The Troy Chemical Corporation property, Newark, New Jersey, is an active industrial chemical facility. Operations began prior to 1892. The site is drained by the man-made Pierson's Creek in the center of the property. The Creek drains to the south and discharges into Newark Bay. Pierson's Creek was originally part of a private drainage system, constructed in the mid-1800s. From 1892 to 1931 the Heller and Merz Company manufactured dyes. Troy Chemical Corporation owned and operated on the property from 1953 to the present. All of Troy's process wastewaters, including mercury, were discharged untreated into Pierson's Creek until 1965.

Troy had several operations until about 1965 when a closed-pipe mercury treatment system was built. In 1968 mercury containing products were manufactured. Mercury recovery stills were in operation from 1974 to 1980. Fungicides were manufactured beginning in approximately 1976. This facility continues to manufacture numerous chemicals.

In 1977 Troy installed a containment wall to prevent spilled/leaking materials from migrating into Pierson's Creek. However, numerous inspections revealed cracks and holes that allowed plant wastewater to discharge directly to the Creek. The firm had many violations of Passaic Valley Sewage Commission (PVSC), and NJ Department of Environmental Protection regulations and permit conditions. Even upgradient offsite soils contained mercury concentrations (140-190 ppm Hg) approaching those considered dangerous. (The New Jersey DEP non-residential direct contact cleanup criterion is 270 ppm.) From January to May 1979 the PVSC collected numerous 24-hour composite samples of the Troy liquid waste. Analyses of these samples indicated that the company was discharging about 330 pounds (= 150 kg) of mercury per day to the PVSC sewage system. Concentrations in onsite sediments (>22,000 and >11,000 ppm Hg) indicate continuous discharges and/or spills of insoluble forms of mercury over several decades.³⁵

Soil contained mercury at concentrations >2,000 ppm in 1988. Shallow ground water (without particulates) contained mercury at approximately ground water quality standards (1 ppb) and deep ground water was <1 ppb Hg from 1996 to 1999.³⁶

A Ventron/Velsicol chemical processing plant was operated from 1929 to 1974 at a 40-acre site within the Hackensack Meadowlands in the boroughs of Wood-Ridge and Carlstadt, New Jersey. The site is bordered on the east by Berry's Creek, on the south by Diamond Shamrock/Henkel Ditch and Nevertouch Creek, and on the north by Ethel Boulevard and a railroad track. Various firms constructed and operated a mercury processing plant, and manufactured mercury products on the site. The plant manufactured red and yellow oxide of mercury, phenyl mercuric acetate, and reclaimed mercury from amalgams, batteries, impure mercury, etc. Ventron operated the plant until it was closed in 1974.³⁷

After 1960 a 19-acre portion of the site near Berry's Creek was used as a dumping area for various mercury-containing materials.³⁸ Much of this area is intertidal filled land. As recently as September 2000 most mercury concentrations in surface soils and sediments of the site ranged from hundreds to thousands of ppm. Soils up to 600' outside a site boundary ranged from 2 to 550 ppm Hg. Mercury vapor concentrations at several sites ranged up to 3,300 ng/m³, with potential effects on >100,000 residents of several municipalities. Conservatively, >160 tons of mercury were dumped on 19 acres with no attempt at containment.³⁹

³³ NJ DEP, LCP Chemicals, Inc. Superfund Site

³⁴ NJ DEP, Final Draft Site Inspection, LCP Chemicals, Inc.

³⁵ Troy Chemical Company, Inc.

³⁶ ELM. (2000). Troy Chemical Company Remedial Investigation Report.

³⁷ U.S. EPA Region 2. (2000).

³⁸ Z. Kelman. Personal communication (2001).

Smelting and refinement of copper and lead emitted large quantities of mercury to air and water until the first emission controls³⁹ in the 1970s. At least four secondary copper smelters were located close to New York/New Jersey Harbor in recent decades (Barth Smelting and Refining Co. and Kearney Smelting and Refining Co. in northern New Jersey, Nassau Smelting and Refining Co. on Staten Island, and Belmont Smelting Co. in Brooklyn). Metal refineries were also concentrated on the margins of the Harbor. During 1899 these firms produced 117,000 (short) tons of refined copper (39% of total U.S. production). As recently as 1970 New York/New Jersey Harbor still accounted for 25% of U.S. copper refining. The same area in 1899 produced 54% of all U.S. copper sulfate production (>7,000 tons). At least five firms continued to manufacture copper sulfate in recent years. Lead refining was also an important regional industry, resulting in large mercury emissions. Two lead refineries (Balbach Metals, built in 1883 in Newark, NJ, and American Smelting and Refining Co., built in 1898 in Perth Amboy) produced 244,000 tons of refined lead in 1899, or 33% of the U.S. production in that year. Additional refineries were located in northern New Jersey. Several secondary lead smelters are still operating in and near Perth Amboy.⁴⁰

Ayres *et al.*⁴¹ have estimated that more than five million tons of mercury were lost to the Hudson-Raritan drainage basin from 1880 through 1980. We have not attempted to quantify emissions from individual firms, but it is possible to estimate lower bounds on regional mercury waste. Until 1980 or so nearly all Hudson-Raritan industries with large mercury emissions were sited near the New York/New Jersey Harbor. Assuming conservatively that 80% of the emissions were near the Harbor margins, this would have amounted to > four million tons of mercury from 1880 through 1980.

Although, it is not possible to apportion quantitatively the Hg losses to the various environmental compartments (i.e., atmosphere, soils, sediments, and waters), it is likely that the local accumulation was >10,000 kg per year for 100 years, or nearly the mass of mercury used annually by regional industry on average from 1980 to the present. (About 50,000 kg of mercury is the regional annual mass used – not emitted -- by modern industry [Table 1]. Industrial emissions have been reduced greatly in recent years.)

The flux of mercury from the Harbor margins to sediments and the ocean is exceptionally complex, particularly over the past century. As stated, the uncertainties in this process do not permit a quantitative estimation of flux. However, residual signatures in mercury distribution do indicate that the concentrated industry on the shores of Newark Bay and the Arthur Kill were large mercury sources, relative to the rest of New York/New Jersey Harbor. The chemistry of Harbor surficial sediments (upper 2 cm) was analyzed in detail in 1993-94. One hundred sixty eight samples of the Harbor sediments were allocated with a stratified random strategy so that confidence limits could be placed on sample statistics.⁴² Using data from Adams *et al.* (1998), mean mercury concentration in Newark Bay surficial sediments (including the Arthur Kill) was 2.6 ± 0.58 ppm whereas the mean concentration in the rest of the Harbor was only 0.62 ± 0.19 ppm.

About 8,800 kg of mercury remains in the upper two centimeters of the Harbor, excluding coastal sediments. The total mass in sediments to depths of industrial influence is obviously much larger. Also substantial quantities of the Harbor mercury will have been flushed to the New York Bight Apex and western Long Island Sound.⁵⁰ Although today's mercury concentrations have been measured at only a few locations in soils close to the Arthur Kill and Newark Bay, much of this area remains heavily contaminated with mercury (and other toxicants). It would be surprising if less than 50,000 kg of mercury remained in these soils. Moreover, the remobilization, delivery flux, and availability of this mercury to the biosphere is a significant environmental and public health concern.

³⁹ URS Corporation. Materials Balance and Technology Assessment of Mercury ...

⁴⁰ Ayres and Ayres, *Accounting for Resources* 2, Ch. 8.

⁴¹ Ayres *et al.* *An Historical Reconstruction of Major Pollutant Levels in the Hudson-Raritan Basin 1880-1980*, See also Table 8-10 of Ayres and Ayres (1999).

⁴² Adams *et al.* (1998), Sediment Quality of the NY/NJ Harbor System.

D. Cultural and Religious Uses⁴³

According to the New Jersey Governor's Task Force on Mercury, a little understood, but possibly consequential source of mercury stems from cultural and religious uses in Afro-Caribbean and Latin American ethnic communities.⁴⁴ Mercury is sold in many *botanicas*⁴⁵ throughout the region, for use in various religious ceremonies. A survey of 115 *botanicas* in 13 cities in the United States and Puerto Rico found that 86% sold mercury.⁴⁶ Johnson's survey of 203 Caribbean and Latin American adults in the New York City area demonstrated that 44% of Caribbean and 27% of Latin American respondents reported using mercury.⁴⁷

Mercury is usually sold in the form of capsules that can contain 8 or 9 grams (0.3 oz.) of mercury.⁴⁸ Mercury is often recommended to store patrons as a means to bring luck in love, money or health, and to ward off evil.⁴⁹ Most who buy the mercury carry it with them in a sealed pouch (49%) or in a pocket (32%) as an amulet. Close to 1/3 of respondents reported sprinkling mercury in their homes. C. Johnson found that 64% of mercury users in his study reported throwing mercury in the garbage, while 27% flushed it down the toilet and 9% threw it outdoors.⁵⁰ Mercury poisoning has been documented in Mexican-American infants who have been fed mercury as a folk remedy for gastroenteritis.⁵¹ Usage and sales estimates vary significantly. Although existing data do not permit precise estimates of usage, order-of-magnitude estimates are possible from estimated sales in New Jersey and scaling population appropriately. A 1995 survey of 35 *botanicas* reported that they sell approximately 100-300 capsules each day.⁵² The U.S. census estimated 720,000 Hispanics or Latinos in New Jersey in 1995. The New Jersey mercury task force estimated that 13,000-38,000 capsules were sold in 1995, containing a total of 100-350 kg of mercury.⁵¹ The Hispanic or Latino population of the Hudson/Raritan Basin reached three million in 2000.⁵³ Assuming similar per capita purchases of capsules over space and time, recent census estimates of population of Hispanic or Latino origin in the Hudson/Raritan basin, indicate that 400-1400 kg (880-3,100 lbs) of mercury were purchased for religious purposes during the year 2000 (100-350 kg • 3 Mil/720K Hispanics or Latinos). Purchases within the Basin in 1980 and 1990 were roughly proportional to Hispanic or Latino population within the Basin (1980 = 1,701,814 people, 200-800 kg Hg purchased; 1990 = 2,370,794 people, 300-1200 kg Hg purchased).

These usage estimates, although very imprecise, are small relative to nearly all sources of mercury, and quantities used in consumer products. Indeed, the quantities used for religious and cultural purposes are small fractions of the total mass incorporated in consumer products (Table 6). However, continuing direct exposures to mercury through these practices are potential health hazards.

IV. MERCURY FLOWS AND CYCLING: ATMOSPHERIC, RIVERINE AND WASTEWATER MERCURY FLUXES RELATING TO NY/NJ HARBOR

⁴³ The text and data on mercury usage in religion is adapted from the NJ Governor's Task Force on Mercury.

⁴⁴ A. Pinn (1998), *Varieties of African American Religious Experience* (Minneapolis: Fortress Press).

⁴⁵ *Botanicas* are stores that specialize in herbal remedies and religious items used in Santeria (Lukumi), Voodoo, and Espiritismo.

⁴⁶ A. Wendroff (1990), "Domestic Mercury Pollution," *Nature* 347: 623; C. Johnson (1999), "Elemental Mercury Use in Religious and Ethnic Practices in Latin American and Caribbean Communities in New York City," *Population and Environment* 20 (5): 443-453.

⁴⁷ C. Johnson (1999), "Elemental Mercury Use"

⁴⁸ L.H. Zayas and P.O. Ozuah (1996), "Mercury Use in Espiritismo: A Survey of Botanicas," *American Journal of Public Health* 86 (1): 111-112.

⁴⁹ Zayas and Ozuah, "Mercury Use in Espiritismo"

⁵⁰ C. Johnson (1999), "Elemental Mercury Use"

⁵¹ Geffner and Sandler (1980)

⁵² Zayas and Ozuah, "Mercury Use in Espiritismo"

⁵³ www.census.gov; census 2000 data now available, race: Hispanic or Latino, county. (U.S. Census data combines people of Hispanic and Latino backgrounds.)

The atmosphere plays a primary role in cycling natural and pollution derived Hg over local, regional, and global scales (Mason et al., 1994; Fitzgerald et al., 1998). A significant portion, for example, of the riverine Hg entering NY/NJ Harbor is anthropogenic and due principally to mobilization of Hg from the watershed. Moreover, much of the Hg in the watershed has been deposited via atmospheric processes. This Hg, as noted, is principally anthropogenic, and derived from extra-and intra-regional sources. Recent studies suggest that about 25-30% of the Hg deposited in a watershed can be leached into rivers and lakes (Swain et al., 1994; Lamborg et al., 2000). This information can be used to estimate the potential watershed contribution from atmospherically derived Hg in fluvial transport to the Harbor. Environmental and management insights are gained when this predicted Hg flux is compared to the Hg flux based on measured Hg levels and stream flow for the Hudson. For example, this biogeochemical scaling for the combined Hudson/Raritan/Passaic/Hackensack watersheds allows an initial assessment of the importance of atmospheric Hg sources to the current accumulation of Hg in the sediments of NY/NJ Harbor. This evaluation also yields a scale for assessing the significance of Hg contributions from sources other than the atmosphere.

As stated earlier, “our goals are to provide: 1. the proper magnitude for the Hg flows relating to the NY/NJ Harbor, and 2. a firm scientific basis for assessing the scale and impact of pollution-related Hg. The current database is inadequate for more refined approaches or simulations. Thus, and in view of the weaknesses in the available biogeochemical and environmental information for Hg in the region, reasonable assumptions and simplifications are made. Our approach provides a first-order estimate for a mass balance of Hg transport and deposition within NY/NJ Harbor. We are laying the framework for a realistic assessment of Hg sources, cycling, behavior and fate in the Harbor.”

In this work, we use the total mean annual river flow into NY/NJ Harbor. Beardsley and Boicourt (1981) estimate this transport at $873 \text{ m}^3 \text{ sec}^{-1}$, which represents the streamflow from the Hudson, Raritan, Passaic, and Hackensack rivers. We estimate this average annual flow at $708 \text{ m}^3 \text{ sec}^{-1}$ using USGS gauge data from 1973-1994 (information available at <http://waterdata.usgs.gov/nwis-w/US>). A watershed adjustment was made for the streamflow of the Hudson (1.58 X the Green Island Station values) into the Harbor. The USGS summary indicates that the Hudson contributes about 90% of the annual flow, or ca. $650 \text{ m}^3 \text{ sec}^{-1}$, while the Raritan, and Passaic/Hackensack transport provides about 10%. There is an East River contribution that is difficult to assess. Blumberg and Pritchard (1997) suggest that a $310 \text{ m}^3 \text{ sec}^{-1}$ flow enters the East River from Long Island Sound, and about $45 \text{ m}^3 \text{ sec}^{-1}$ enters from wastewater treatment facilities. Some of the East River transport is diverted into the Hudson through the Harlem River. The average total annual riverine flow ranges from $1063\text{-}1228 \text{ m}^3 \text{ sec}^{-1}$ using $355 \text{ m}^3 \text{ sec}^{-1}$ for the East River portion. We chose the mean value of $1150 \text{ m}^3 \text{ sec}^{-1}$. The USGS analysis suggests that the annual variation for the streamflow is about 20%. Thus, the streamflow range associated with the prior work is comparable to expected variability. Finally and to simplify the presentation of riverine Hg flux calculations, we use the Hudson* to represent the total riverine contribution from the Hudson, Raritan, Passaic/Hackensack, and East Rivers into the Harbor.

In addition, we have assumed that the levels of Hg determined for the Hudson River by Sañudo-Wilhelmy and Gill (1998) are equivalent to that expected in the Raritan, Passaic, and the Hackensack. Thus, the annual riverine Hg flux to the Harbor is equal to the total riverine annual streamflow (i.e., Hudson*) times the measured concentrations of Hg in the Hudson river.

Given the basis for the streamflow, we assume in our analysis that 25% of the combined annual atmospheric mercury deposition from local and long-range sources is contributed to the Hudson* from the watershed. This predicted value greatly exceeds the empirical estimates of the dissolved Hg river flux. Indeed, our analysis suggests that only 15% of the atmospherically derived Hg that enters the river from the watershed is transported as dissolved Hg to the Harbor. The remainder or 85% accumulates in riverborne particles and in the sediments. This comparison suggests the likelihood of substantial accumulation of atmospherically derived Hg in the river sediments. We emphasize that such sequestered Hg is available for biogeochemical interactions and transformations. Such reactions include the biologically mediated synthesis of the very toxic methylHg species (Compeau and Bartha, 1985; Winfrey and Rudd, 1990; Gilmour and Henry, 1991).

Finally and as outlined in the following sections, our simulation indicates that atmospherically related Hg inputs can account for the observed Hg levels in both the dissolved and particulate phases in the Hudson*. Therefore, there would be a decline in Hg entering the rivers following a decline in atmospheric deposition of Hg. Further, and since intra- and extra-regional atmospheric Hg sources of Hg are similar, in-basin (i.e., localized) reductions in Hg emissions will lead to a decrease in Hg entering the rivers and a concomitant reduction in Hudson* Hg discharges into NY/NJ Harbor. In addition, and we stress, methylHg production in the rivers and watersheds should be reduced as local mercury emissions are diminished.

A. Tropospheric Hg Burden

The tropospheric mercury burden for the Hudson* basin is 380 kg. This estimate is based on an atmospheric Hg concentration of $1.8\text{g}/\text{km}^3$, an area of the watershed at $3.4 \times 10^4 \text{ km}^2$ and an isobaric tropospheric volume of $21 \times 10^4 \text{ km}^3$ (Lamborg et al., 1999). Most of this Hg is in the elemental form, the majority of which is unaffected as it passes through the region, and into the global circulation. This annual advective atmospheric flux of elemental mercury, while large and uncertain, is not a factor in our evaluation. Rather, we are focused on the reactive components of the atmospheric cycling of Hg which deposit Hg in the region. Therefore, the annual Hg transport through the regional airshed is considered to be unchanged and can be ignored in our first-order simulation.

B. Atmospheric Hg Deposition and Hudson* Fluxes of Mercury

Extra-regional and longer-range sources. Atmospheric deposition of mercury associated with extra-regional and longer-range sources was estimated using a tested and published technique. In this method, mercury deposition is indexed to the better-known depositional flux of the atmospheric natural radiotracer, ^{210}Pb , to predict a mercury depositional flux to the Hudson* River Watershed of $360 \pm 60 \text{ kg yr}^{-1}$ (Lamborg et al., 2000; Table 4). Using the estimate for the leaching of Hg from a watershed (@ 25% of the atmospheric depositional flux), a mercury input to the river of $90 \pm 15 \text{ kg yr}^{-1}$ is obtained.

Table 4.
Mercury Flux from Extra-Regional Sources to the Hudson* River and NY/NJ Harbor

^{210}Pb Deposition to Hudson* Watershed	$6 \pm 1 \times 10^{15} \text{ mBq yr}^{-1}$
Predicted Hg Depositional Flux to Hudson* Watershed (<i>extra-regional longer range sources</i>)	$360 \pm 60 \text{ kg yr}^{-1}$
Assuming 25% Loss of Hg from Watershed to River, Predicted River Flux	$90 \pm 15 \text{ kg yr}^{-1}$
Average Hudson* Streamflow (ca. 20% average variation)	$1150 \text{ m}^3 \text{ sec}^{-1}$

Predicted Hudson* Total Hg concentration	13 pM*
Measured dissolved Hg _D range (1-7 pM) ⁵⁴ ; Average	4±1 pM
Particulate Hg _P range (16-129 pM) ⁵⁵ ; Average	49±33 pM
*pM (1 picomolar = 200 x 10 ⁻¹² g L ⁻¹ or 2 x 10 ⁻¹⁰ kg m ⁻³)	

As summarized in Table 4, and using a mean total stream flow of $3.6 \times 10^{10} \text{ m}^3 \text{ yr}^{-1}$ an average Hg_{total} concentration of **13 pM** in the Hudson* is expected from the mercury entering via watershed runoff of extra-regionally derived Hg. The ²¹⁰Pb indexing and the leaching forecast can be compared with recent measurements in the Hudson River.⁵⁶ Thus, the extra-regionally derived Hg yields a predicted level that is 3 times larger than the average dissolved Hg_D of 4±1 pM inferred from the Sañudo-Wilhelmy and Gill (1998) measurements. As discussed in the next section, the transfer to the river of atmospherically derived is larger since depositional fluxes of Hg to the watershed from in-region sources must be included. This difference would suggest that watershed derived Hg is being sequestered and accumulating in river sediments. This inference is supported by the levels (49±33 pM) of particulate associated (Hg_P) measured at the Battery and upriver by Sañudo-Wilhelmy and Gill (Table 4).

Much of the Hg_P measured in the Hudson probably represents resuspended material. Indeed, it is likely most of the particulate Hg delivered to the River will eventually deposit in the River. The uncertainty, however, argues for a conservative approach, which yields an order of magnitude range for the Hg flux from the Hudson*. That is, a minimum riverine Hg flux to the Harbor assumes no particulate Hg transport, while the maximum flux assumes all of the measured riverborne dissolved and particulate phases of Hg enter the Harbor. Thus, using an annual riverine flow of $3.6 \times 10^{10} \text{ m}^3$, the estimated average range for the transport of Hg from the Hudson* is 29±7 kg yr⁻¹ to 381±245 kg yr⁻¹. Conservatively, and since the variation in the total Hg flux overlaps the dissolved Hg transport estimate at the two sigma limit, 381±245 kg yr⁻¹ is employed for the annual mean fluvial input of Hg to NY/NJ Harbor.

Deposition: local/regional sources. As summarized in Table 2, fossil fuel combustion (1997 and 2000) yields an annual Hg flux to the atmosphere of 770 kg with an uncertainty of 40%. Municipal solid waste (MSW) incineration provides an additional $140 \pm 70 \text{ kg}$ of Hg per year. Therefore total Hg emissions from these sources are $910 \pm 315 \text{ kg}$ per year. The chemical composition of these Hg emissions is crucial to estimating the amounts of Hg that are likely to deposit within the region. Unfortunately, Hg emissions/speciation studies are much too sparse to provide a refined estimate. Here we assume that 40% of the Hg emitted from fossil fuel combustion processes and MSW incineration is composed of reactive gaseous forms. Such reactive species have an extremely short lifetime in the atmosphere and will deposit within the Hudson* basin (EPA, 1997 report). The remaining 60% is assumed to enter the global cycle as the elemental form.

We estimate that local Hg deposition of Hg emitted from in-basin fossil fuel combustion processes and MSW incineration will be 40% of the total flux ($910 \pm 315 \text{ kg}$ per year).

This depositional flux is about $365 \pm 125 \text{ kg}$ per year and quite significant. It is nearly identical to the amount estimated for in-basin Hg deposition associated with extra-regional sources ($360 \pm 60 \text{ kg yr}^{-1}$).

In Table 5, mercury deposition predictions based on local/regional Hg atmospheric sources as well as the extra-regional and longer range contributions (cf; Table 4) are listed. In addition, the tabulation includes the predicted Hudson* Hg concentration from atmospheric deposition and leaching from the watershed, the mean Hudson river mercury flux for dissolved and particulate Hg derived from the analytical data of Sañudo-Wilhelmy and Gill, and the mean concentration found in the Connecticut River by the U. of Connecticut program (i.e., Fitzgerald and collaborators).

⁵⁴ Sañudo-Wilhelmy, S.A. and Gill, G.A., 1998, and 1999.

⁵⁵ Ibid.

⁵⁶ S.A. Sañudo-Wilhelmy and G.A. Gill (1999), "Impact of the Clean water Act on the Levels of Toxic Metals in Urban Estuaries: The Hudson River Estuary Revisited," *Environ. Sci. Technol.* 33: 3477-3481.

Geochemical and Anthropogenic Implications. Firstly, the predicted fluvial fluxes associated with direct deposition from local/regional Hg atmospheric sources are comparable to the longer range contributions. Indirect input of Hg to the rivers (e.g., via runoff) from local/regional Hg atmospheric sources is about $91 \pm 31 \text{ kg yr}^{-1}$. The contribution from extra-regional and longer-range sources is $90 \pm 15 \text{ kg yr}^{-1}$. Secondly, the predicted Hudson River total Hg concentration is 25 pM, which 6 times the 4 pM value for dissolved Hg inferred from Sañudo-Wilhelmy and Gill measurements. Therefore, and quite importantly, approximately 85% of the Hg entering the Hudson/Raritan from the watershed is predicted to be sequestered in particulate matter and/or sediments. Thirdly, and as noted, some portion of the sequestered Hg is available for biogeochemical interactions and transformations. Such reactions, as noted, include the biologically mediated synthesis of the very toxic methylHg species.. Fourthly, the predicted watershed delivery of atmospherically derived Hg yields an average Hg level in the Hudson similar to the mean total Hg values ($53 \pm 33 \text{ pM}$; 90% Hg_p) reported by Sañudo-Wilhelmy and Gill. The contribution of resuspended sediments containing Hg to the riverine Hg_p is likely to be significant.

Table 5.
Predicted Hudson* Hg Flux (Comparison to other work)

Assuming 25% Loss of Hg from Predicted Extra-Regional Hg Depositional Flux to Hudson* Watershed ($360 \pm 60 \text{ Kg yr}^{-1}$) yields a River Flux	$90 \pm 15 \text{ kg yr}^{-1}$
Assuming 25% Loss of Hg from Predicted Localized Hg Depositional Flux to Hudson* Watershed ($365 \pm 125 \text{ kg yr}^{-1}$) yields a River Flux	$91 \pm 31 \text{ kg yr}^{-1}$
Average Hudson* Stream flow	$1150 \text{ m}^3 \text{ sec}^{-1}$ ($3.6 \times 10^{10} \text{ m}^3 \text{ yr}^{-1}$)
Predicted Hudson* Total Hg concentration	25 pM
Measured dissolved range 1-7 pM ⁵⁷ ; average	4\pm1 pM
Particulate Hg_p range (16-129 pM)⁵⁸ ; Average	49\pm33 pM
Measured Hg_{Total} in Connecticut River	22 \pm 4 pM⁵⁹
Predicted Hudson* Hg Flux (meas. conc. x mean flow) for dissolved and total Hg (particulate and dissolved phases).	29 \pm 7 kg yr^{-1} dissolved Hg, and 381 \pm 245 kg yr^{-1} total Hg.

However, and because resuspension is difficult to quantify, another interpretation can be offered which argues that essentially all of the Hg entering the river via deposition from localized and extra-regional Hg atmospheric sources is transported seaward into the Harbor. As outlined above, the estimated average range for the transport of Hg from the Hudson is 29 \pm 7 kg yr^{-1} for dissolved Hg to 381 \pm 245 kg yr^{-1} for the dissolved plus particulate associated Hg. Fifthly, independent of interpretation, and a quite significant implication, this analysis indicates that there would be decline in Hg entering the rivers following a decline in atmospheric deposition of Hg. Moreover, in-basin (i.e., localized) reductions in Hg emissions will lead to a decrease in the Hg flux from the watershed into the fluvial flow seaward. Thus, there should be a concomitant reduction in Hg discharges from the Hudson* (i.e., Hudson, Raritan, Passaic and Hackensack) to NY/NJ Harbor. Sixthly, methylHg production in the rivers and watersheds should be reduced as local mercury emissions are diminished. Seventhly, similar improvements are implied for a reduction in the contribution from extra-regional sources.

C. Emissions from Soil

⁵⁷ Sañudo-Wilhelmy, S. and Gill, G. (1999), "Impact of the Clean Water Act on the Levels of Toxic Metals in Urban Estuaries. The Hudson River Estuary Revisited." *Environ. Sci. Technol.* 33: 3477-3481.

⁵⁸ Sañudo-Wilhelmy, S. and Gill, G. (1998). "Phase-speciation of Toxic Metals in the Hudson River Estuary." *Final Report to the Hudson River Foundation.*

⁵⁹ Fitzgerald, et al. (2000) , "Mercury Emissions and Cycling in the Coastal Zone," *J. Environ. Sci.* 12: 92-101; Rolfhus and Fitzgerald (2001), "The Spatial/Temporal Distribution and Evasion of Mercury Species in Long Island Sound, CT," *Geochim. Cosmochim. Acta*, 3: 407-418.

Average emissions of mercury from soils to atmosphere have been estimated $9.7 \pm 3.9 \text{ g km}^{-2} \text{ yr}^{-1}$. Scaling up to include the Hudson* watershed area, we arrive at an estimate of $330 \pm 132 \text{ kg yr}^{-1}$.⁶⁰ Uncertainty is estimated again at $\pm 40\%$.

D. Wastewater Treatment Facility/Sewage Inputs

The annual mercury input to the Harbor is estimated from the volume of sewage/wastewater discharged to the Harbor and the concentration of total mercury in the sewage. The annual estimated volume of wastewater/sewage delivered to the Harbor from Water Pollution Control Plants (WPCP) is $3 \times 10^{12} \text{ L}$ (ca. 65 % from NY).⁶¹ Most measurements of mercury in New York and New Jersey sewage/wastewater were made before clean methods were in use. Thus, these early estimates are highly unreliable. The estimated total mercury concentration in New York-area WPCP effluents is presumed to be similar to that measured in Connecticut waste treatment facilities: $76 \pm 27 \text{ pM}$ ($15 \pm 5.4 \text{ ng/L}$, $n=22$). Thus, the estimated annual input of total mercury from sewage is $(3 \times 10^{12} \text{ L}) \times (76 \pm 27 \text{ pM}) = 46 \pm 16 \text{ kg}$.⁶²

For comparative purposes and summarized in Table 5., the estimated annual Hg input to the Harbor from the Hudson* is 29 ± 7 to $381 \pm 245 \text{ Kg}$. The larger value, $381 \pm 245 \text{ Kg yr}^{-1}$, as described, will be used because it is likely that the Hudson* input to the Harbor is contained within the uncertainty.

E. Direct Net Atmospheric Hg Exchange

There is a annual net Hg input of $7 \pm 3 \text{ kg}$ to the atmosphere from the Harbor [$(11 \pm 4) - (4 \pm 1) \text{ kg}$]. The estimated wet depositional flux of mercury to the NY/NJ Harbor is $4 \pm 1 \text{ kg yr}^{-1}$.⁶³ This is obtained by scaling from a recent estimate of $26 \pm 7 \text{ kg/year}$ for Long Island Sound.⁶⁴ Similarly, the estimate for Hg emissions (as elemental Hg) is $11 \pm 4 \text{ kg yr}^{-1}$.

F. Net Hg Tidal Exchange

The tidal velocities are high and therefore the advective Hg fluxes are large. However, a conservative estimate of net water exchange in the NY/NJ Harbor would yield water residence time as long as 10 days for a Harbor volume of $1.9 \times 10^{12} \text{ L}$ (i.e., 5 m average depth). However, given the paucity of Hg data, especially the partitioning between the dissolved and particulate phases, an estimate for the net transfer of Hg associated with tidal exchange is too uncertain to include. This is a research need.

G. Total annual input of Hg to the Harbor

⁶⁰W.F. Fitzgerald and R.P. Mason (1996), "The Global Mercury Cycle: Oceanic and Anthropogenic Aspects," in *Regional and Global Mercury Cycles: Sources, Fluxes and Mass Balances*, W. Baeyens, O. Vasiliev and R. Ebinghaus, eds., (The Netherlands: Kluwer Academic Publishers), pp. 85-108.

⁶¹ Interstate Sanitation Commission (1997), *Annual Report* (NY: ISC).

⁶² See Fitzgerald, et al. (2000), "Mercury Emissions and Cycling in the Coastal Zone," *J. Environ. Sci.* 12: 92-101; Rolfhus and Fitzgerald (2001), "The Evasion and Spatial/Temporal Distribution of Mercury Species in Long Island Sound, CT-NY," *Geochim. Cosmochim. Acta*, 65: 407-408 This value is likely to change.

⁶³ Fitzgerald, et al. (2000), "Mercury Emissions and Cycling in the Coastal Zone," *J. Environ. Sci.* 12: 92-101.

⁶⁴Ibid.; Rolfhus and Fitzgerald (2000), The Spatial/Temporal Distribution and Evasion of Mercury Species in Long Island Sound, CT: *Geochim. Cosmochim. Acta*,

As noted, the net tidal Hg transfer is uncertain. Thus, the total annual input of Hg to the Harbor is 381 ± 245 kg from fluvial inputs (Hudson*), and 46 ± 16 kg (wastewater/sewage) minus 7 ± 3 kg (net atmospheric emissions). This net annual input is 420 ± 250 kg.

This input estimate is much less than the 3,100 kg estimated by the Harbor Estuary Program or the 11,700 kg estimated by HydroQual, Inc. Both estimates are evidently much too large for current conditions.⁶⁵

H. Deposition to Harbor Sediments

The annual deposition of mercury to Harbor sediments can be estimated from the rate of fine-particle accumulation in the Harbor ($1.5 \pm 0.5 \times 10^{12}$ g) and the partitioning of mercury between particulate and dissolved phases in seawater.⁶⁶ The mean mercury concentration in Harbor waters is 3.91 ± 0.86 pM ($0.78 \pm 0.17 \times 10^{-12}$ g/g).⁶⁷ The distribution coefficient, K_D , is used to quantify the partitioning of mercury between particulate and dissolved phases in seawater: $K_D = (\text{Hg [g/g] particles})/(\text{Hg [g/g] water})$.

A mean K_D for Hg = ($10^{5.6 \pm 0.3}$) has been estimated from 21 surface water samples from the Connecticut River mixing zone, the East River, western and central Long Island Sound.⁶⁸ Therefore,

$$\begin{aligned} (\text{Hg [g/g] particles}) &= K_D \times (\text{Hg [g/g] water}) \\ &= 10^{5.6 \pm 0.3} \times 0.78 \pm 0.17 \times 10^{-12} \text{ g/g} \\ &= 310 \pm 150 \times 10^{-9} \text{ g/g.} \end{aligned}$$

So, we estimate the annual flux of mercury to Harbor sediments as:

$$\begin{aligned} (\text{Hg [g/g] particles} \times 1.5 \times 10^{12} \text{ g}) \text{ is} \\ 310 \pm 150 \times 10^{-9} \text{ g/g} \cdot 1.5 \pm 0.5 \times 10^{12} \text{ g} = 470 \pm 280 \text{ kg.} \end{aligned}$$

The concentration of mercury on the fine particles settling out is 310 ± 150 ppb, which is surprisingly close to the 740 ± 140 ppb estimated from 168 randomly selected, direct measurements of surficial sediments (top 2 cm) in the Harbor.⁶⁹ All these samples were taken in late summer of 1993 and 1994.

The preliminary budget for mercury in NY/NJ Harbor shows strikingly good agreement between the Hg deposition flux and our estimated annual inputs. Indeed, the net annual input which is 420 ± 250 kg (from wastewater/sewage and river sources) is effectively the same as our estimate for Hg deposition to the sediments at 470 ± 280 kg. Given the uncertainties, our estimates are well constrained. This agreement suggests that much of the Hg entering from wastewaters and the rivers will deposit in the Harbor. Such an inference is consistent with the partitioning of mercury in aqueous phases where it is principally associated with particulate material. The similarity of Hg inputs and outputs via deposition in the Harbor suggests that net tidal Hg exchange, while uncertain, may not be large.

V. ESTIMATES OF ANNUAL MERCURY FLUX

Table 6.

⁶⁵ NY-NJ Harbor Estuary Program (1996), *Comprehensive Conservation and Management Plan* (NY: US EPA); HydroQual (1991), *Assessment of Pollutant Loadings to the New York-New Jersey Harbor* (Mahwah, NJ: US EPA Contract), Task 7.1.

⁶⁶ C.R. Olsen, et al. (1984), "A Geochemical Assessment of Sedimentation and Contaminant Distribution in the Hudson Raritan Estuary," *NOAA Tech Report* (Rockville, MD: NOS OMS 2).

⁶⁷ Sañudo-Wilhelmy and Gill, "Phase-speciation of Toxic Metals."

⁶⁸ W.F. Fitzgerald (2000), unpublished results.

⁶⁹ D.A. Adams, J.S. O'Connor, and S.B. Weisberg (1998), *Sediment Quality of the NY/NJ Harbor System* (NY: USEPA Region 2), EPA/902-R-98-001, Table E.1.

Annual Mercury Flux to/from the NY/NJ Harbor Watersheds Over Two Decades

SOURCES	1980 1,000 kg/y	1990 1,000 kg/y	2000* 1,000 kg/y
Use in Industrial Operations**	102	27.8	10.6
Emissions from Industrial Operations	<<100	<<28	<<11
Fossil Fuel Combustion	0.77±0.3	0.85±0.33	0.77±0.3
Municipal Solid Waste Incineration			0.14 ± 0.07
Solid Waste Deposited in Landfills			34±17
Soil Emissions			0.33 ± 0.13
INTERMEDIATE RECEPTORS			
Use in Consumer Products**	102-103	28-29	11-12
Surface Waters (Hudson*)			0.38 ± 0.25
Wastewater Treatment Facilities			0.05 ± 0.02
"PERMANENT" RECEPTORS			
Local Atmospheric Deposition			0.37 ± 0.14
Regional Atmospheric Deposition			0.36 ± 0.06
Transport to Harbor via Rivers			0.38 ± 0.25
Transport to Harbor via Wastewater			0.05 ± 0.02
Harbor Deposition on Particles			0.47 ± 0.28

* "Year 2000" estimates are based upon the most recent [1997 to 2000] data.

****The mass of mercury used by industry, and incorporated in products. These are not losses from industrial operations, or from consumer products. The mass of mercury [used] in consumer products is the mass used in industrial operations plus that used for religious and cultural practices.** U.S. Census Bureau, 1980 and 1990 U.S. Bureau of Mines, Minerals Yearbook; Feb 1998 U.S. Geological Survey, 1993-1996 Minerals Yearbook, Minerals and Materials Information. Feb 2000 U.S. Geological Survey, 1994-1998 Minerals Yearbook, Minerals and Materials Information. Downloaded from CD Rom.

Empty cells of Table 6 lack estimates because of exceptionally large uncertainties. All values in Table 6 have sizeable uncertainties, whether they are quantified or not. As suggested, mean confidence limits on the measured estimates are about $\pm 50\%$. It is unlikely that these uncertainties can be reduced significantly. Confidence limits are seldom estimable for statistics of mercury usage/disposal. However, uncertainties in these estimates are also large. "Permanent" receptors in Table 6 is denoted in quotes because these pools of mercury are probably not permanent. For instance, some of the mercury transported to the Harbor is transported to the Atlantic and, eventually enters the atmosphere to become part of atmospheric background. Most of our flux estimates are internally consistent, i.e., appropriate masses balance. For example, our estimated small losses from modern industrial operations correspond with the lack of emissions (to two digits) from 1980 "metallurgical operations".⁷⁰ Also, Ayres and Ayres' estimate of mercury liberated by fossil fuel combustion in 1980 (27 kg) is reasonably close to our 770 kg estimate (Table 6).

In recent years, consumption-related mercury emissions have overwhelmed production-related emissions. This important consideration was originally emphasized by Ayres and Rod (1986). This has become true of nearly all toxicants, not just mercury. Even recognizing the major uncertainties in the flux estimates of Table 6, there is no evidence that production-related wastes of mercury (essentially zero) approach consumption-related emissions. Indeed consumption-related emissions in 2000 are more than three times the estimated mass used in industrial operations during 2000. This seeming disparity is understandable given the large quantities of mercury in decades-old products only recently discarded and dispersed in soil and water. Further, large quantities of mercury continue to be released from old environmental pools such as historical industrial wastes in northern New Jersey and old consumer

⁷⁰ Ayres and Ayres, *Accounting for Resources*, 2. Table 8.10.

products in “dumps” throughout the study area. We note, for example, that the total concentration of mercury in the atmosphere has increased by 2-3 times in the last 100 years.⁷¹ Most of this mercury now in the environment (at least 90% of it) originally came from anthropogenic sources.

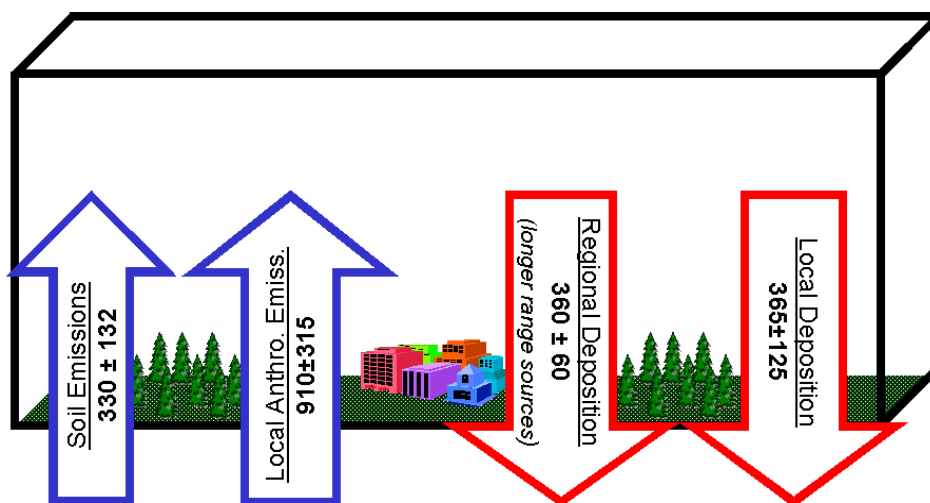


Figure 2. Annual Atmospheric Mercury Flows Hudson/Raritan/Passaic/Hackensack Watershed (kg y^{-1})

VI. MERCURY MASS BALANCES FOR THE HUDSON* WATERSHED AND NY/NJ HARBOR: INDUSTRIAL ECOLOGICAL AND ENVIRONMENTAL IMPLICATIONS

Summaries of the Hg mass balances for the regional atmosphere (Hudson* watershed) and NY/NJ Harbor are presented in the Figures 2 and 3.

Atmospheric Hg Mass Balance

1. Significant in-basin Hg emissions are difficult to detect in the atmosphere given the large tropospheric Hg burden and advection.
2. Hg deposition from extra- and intra-regional sources are large and comparable (total at ca. 725 kg yr^{-1}).
3. Given the expected watershed loss of atmospherically-deposited Hg at 25%, the contribution from the Hudson* watershed to the rivers is about 180 kg yr^{-1} .
4. The estimated total Hg flux from the Hudson* is about 380 kg yr^{-1} . The $> 50\%$ variability suggests that atmospherically-derived Hg may contribute most of the Hg transported via the rivers.
5. There would be decline in Hg entering the rivers following a decline in atmospheric deposition of Hg. In-basin (i.e., localized) reductions in Hg emissions will lead to a decrease in the Hg flux from the watershed into the fluvial flow seaward.
6. MethylHg production in the rivers and watersheds should be reduced as local mercury emissions are diminished.
7. Similar improvements are implied for a reduction in the contribution from extra-regional sources.

NY/NJ Harbor Hg Balance

⁷¹ Mason, R.P., W.F. Fitzgerald, and F.M.M. Morel (1994). "The biogeochemical cycling of elemental mercury: anthropogenic influences." *Geochim. Cosmochim. Acta* 58(15):3191-3198.

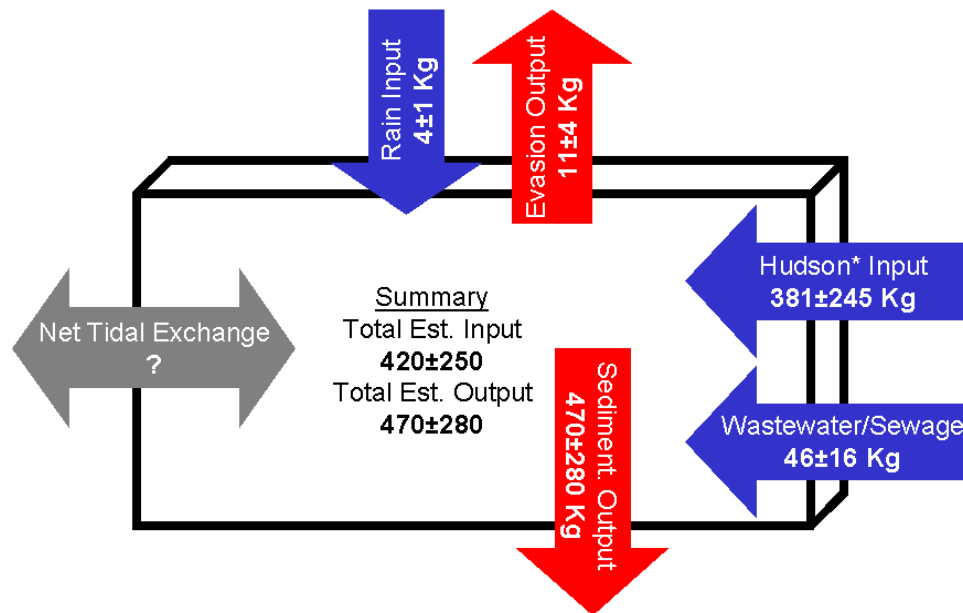


Figure 3. Annual Mercury Flows for NY/NJ Harbor (kg y^{-1})

1. Net Hg input to the Harbor is about $420 \pm 250 \text{ kg yr}^{-1}$. This estimate agrees extraordinarily well with the independent estimate of $470 \pm 280 \text{ Kg yr}^{-1}$ based on the distribution coefficient for Hg and the estimate for sediment accumulation in the Harbor.
2. While the current accumulation of Hg in the Harbor sediments is significantly lower than the early 90's estimates, the anthropogenic contribution is very large. Indeed, 500 kg yr^{-1} Hg accumulation is equivalent to about $1.3 \text{ kg km}^{-2} \text{ yr}^{-1}$. Comparatively, the Hg loadings to Long Island Sound (LIS) are much lower at about $0.08 \text{ kg km}^{-2} \text{ yr}^{-1}$ (Fitzgerald et al., 2000; Rolffhus and Fitzgerald, 2001). The human-related Hg impact on the Harbor is substantial since almost all the Hg in LIS is derived from pollution sources, and these are about 15 times smaller than loading to the Harbor.
3. This mass balance provides a reasonable well constrained working framework. However, little information is revealed about the biogeochemical behavior of Hg in the Harbor. It is essential that speciation (e.g., toxic methylHg, elemental Hg, organo-Hg associations) and geochemical and biological interactions (e.g., methylation/demethylation, uptake, bioaccumulation, transformations) of Hg be examined to assess impact.

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